

12-06-99

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jc662 U.S. PTO
12/03/99

Practitioner's Docket No. 49458

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Box Patent Application
Assistant Commissioner for Patents
Washington, D.C. 20231

jc518 U.S. PTO
09/454316
12/03/99

NEW APPLICATION TRANSMITTAL

Transmitted herewith for filing is the patent application of

Inventor(s): Shien-Chang CHEN, Fu-Shen LIN, Liang-An HSU, Pi-Fwu JANG

WARNING: 37 CFR 1.41(a)(1) points out:

"(a) A patent is applied for in the name or names of the actual inventor or inventors.

(1) The inventorship of a nonprovisional application is that inventorship set forth in the oath or declaration as prescribed by § 1.63, except as provided for in § 1.53(d)(4) and § 1.63(d). If an oath or declaration as prescribed by § 1.63 is not filed during the pendency of a nonprovisional application, the inventorship is that inventorship set forth in the application papers filed pursuant to § 1.53(b), unless a petition under this paragraph accompanied by the fee set forth in § 1.17(i) is filed supplying or changing the name or names of the inventor or inventors."

For (title): CATALYST FOR OXACYLATION AND USE OF THE SAME

CERTIFICATION UNDER 37 C.F.R. 1.10*

(Express Mail label number is **mandatory**.)

(Express Mail certification is **optional**.)

I hereby certify that this correspondence and the documents referred to as attached therein are being deposited with the United States Postal Service on this date December 3, 1999, in an envelope as "Express Mail Post Office to Addressee," mailing Label Number EL299775512US, addressed to the: Assistant Commissioner for Patents, Washington, D.C. 20231.

Susan M. Dillon
(type or print name of person mailing paper)

Susan M. Dillon
Signature of person mailing paper

WARNING: Certificate of mailing (first class) or facsimile transmission procedures of 37 C.F.R. 1.8 cannot be used to obtain a date of mailing or transmission for this correspondence.

***WARNING:** Each paper or fee filed by "Express Mail" **must** have the number of the "Express Mail" mailing label placed thereon prior to mailing. 37 C.F.R. 1.10(b).
"Since the filing of correspondence under § 1.10 without the Express Mail mailing label thereon is an oversight that can be avoided by the exercise of reasonable care, requests for waiver of this requirement will **not** be granted on petition." Notice of Oct. 24, 1996, 60 Fed. Reg. 56,439, at 56,442.

1. Type of Application

This new application is for a(n)

(check one applicable item below)

- Original (nonprovisional)
- Design
- Plant

WARNING: *Do not use this transmittal for a completion in the U.S. of an International Application under 35 U.S.C. 371(c)(4), unless the International Application is being filed as a divisional, continuation or continuation-in-part application.*

WARNING: *Do not use this transmittal for the filing of a provisional application.*

NOTE: *If one of the following 3 items apply, then complete and attach ADDED PAGES FOR NEW APPLICATION TRANSMITTAL WHERE BENEFIT OF A PRIOR U.S. APPLICATION CLAIMED and a NOTIFICATION IN PARENT APPLICATION OF THE FILING OF THIS CONTINUATION APPLICATION.*

- Divisional.
- Continuation.
- Continuation-in-part (C-I-P).

2. Benefit of Prior U.S. Application(s) (35 U.S.C. 119(e), 120, or 121)

NOTE: *A nonprovisional application may claim an invention disclosed in one or more prior filed copending nonprovisional applications or copending international applications designating the United States of America. In order for a nonprovisional application to claim the benefit of a prior filed copending nonprovisional application or copending international application designating the United States of America, each prior application must name as an inventor at least one inventor named in the later filed nonprovisional application and disclose the named inventor's invention claimed in at least one claim of the later filed nonprovisional application in the manner provided by the first paragraph of 35 U.S.C. 112. Each prior application must also be:*

- (i) An international application entitled to a filing date in accordance with PCT Article 11 and designating the United States of America; or*
- (ii) Complete as set forth in § 1.51(b); or*
- (iii) Entitled to a filing date as set forth in § 1.53(b) or § 1.53(d) and include the basic filing fee set forth in § 1.16; or*
- (iv) Entitled to a filing date as set forth in § 1.53(b) and have paid therein the processing and retention fee set forth in § 1.21(l) within the time period set forth in § 1.53(f).*

37 CFR 1.78(a)(1).

NOTE *If the new application being transmitted is a divisional, continuation or a continuation-in-part of a parent case, or where the parent case is an International Application which designated the U.S., or benefit of a prior provisional*

application is claimed, then check the following item and complete and attach ADDED PAGES FOR NEW APPLICATION TRANSMITTAL WHERE BENEFIT OF PRIOR U.S. APPLICATION(S) CLAIMED.

WARNING: *If an application claims the benefit of the filing date of an earlier filed application under 35 U.S.C. 120, 121 or 365(c), the 20-year term of that application will be based upon the filing date of the earliest U.S. application that the application makes reference to under 35 U.S.C. 120, 121 or 365(c). (35 U.S.C. 154(a)(2) does not take into account, for the determination of the patent term, any application on which priority is claimed under 35 U.S.C. 119, 365(a) or 365(b).) For a c-i-p application, applicant should review whether any claim in the patent that will issue is supported by an earlier application and, if not, the applicant should consider canceling the reference to the earlier filed application. The term of a patent is not based on a claim-by-claim approach. See Notice of April 14, 1995, 60 Fed. Reg. 20,195, at 20,205.*

WARNING: *When the last day of pendency of a provisional application falls on a Saturday, Sunday, or Federal holiday within the District of Columbia, any nonprovisional application claiming benefit of the provisional application must be filed prior to the Saturday, Sunday, or Federal holiday within the District of Columbia. See 37 C.F.R. § 1.78(a)(3).*

The new application being transmitted claims the benefit of prior U.S. application(s). Enclosed are ADDED PAGES FOR NEW APPLICATION TRANSMITTAL WHERE BENEFIT OF PRIOR U.S. APPLICATION(S) CLAIMED.

3. Papers Enclosed

A. Required for Filing Date under 37 C.F.R. 1.53(b) (Regular) or 37 C.F.R. 1.153 (Design) Application

11 Pages of Specification (including cover sheet)
3 Pages of Claims
0 Sheets of Drawing

Formal
 Informal

B. Other Papers Enclosed

1 Pages of Abstract
 Other

WARNING: *DO NOT submit original drawings. A high quality copy of the drawings should be supplied when filing a patent application. The drawings that are submitted to the Office must be on strong, white, smooth, and non-shiny paper and meet the standards according to § 1.84. If corrections to the drawings are necessary, they should be made to the original drawing and a high-quality copy of the corrected original drawing then submitted to the Office. Only one copy is required or desired. For comments on proposed then-new 37 C.F.R. 1.84, see Notice of March 9, 1988 . . . (1990 O.G. 57-62).*

NOTE: *"Identifying indicia, if provided, should include the application number or the title of the invention, inventor's name, docket number (if any), and the name and telephone number of a person to call if the Office is unable to match the drawings to the proper application. This information should be placed on the back of each sheet of drawing a minimum distance of 1.5 cm. (5/8 inch) down from the top of the page." 37 C.F.R. 1.84(c).*

(complete the following, if applicable)

The enclosed drawing(s) are photograph(s), and there is also attached a "PETITION TO ACCEPT PHOTOGRAPH(S) AS DRAWING(S)." 37 C.F.R. 1.84(b).

4. Additional Papers Enclosed

Preliminary Amendment
 Information Disclosure Statement (37 C.F.R. 1.98)
 Form PTO-1449
 Citations
 Declaration of Biological Deposit
 Submission of "Sequence Listing," computer readable copy and/or amendment pertaining thereto for biotechnology invention containing nucleotide and/or amino acid sequence.
 Authorization of Attorney(s) to Accept and Follow Instructions from Representative
 Special Comments
 Other:

5. Declaration or Oath

NOTE: A newly executed declaration is not required in a continuation or divisional application provided the prior nonprovisional application contained a declaration as required, the application being filed is by all or fewer than all the inventors named in the prior application, there is no new matter in the application being filed, and a copy of the executed declaration filed in the prior application (showing the signature or an indication thereon that it was signed) is submitted. The copy must be accompanied by a statement requesting deletion of the names of person(s) who are not inventors of the application being filed. If the declaration in the prior application was filed under § 1.47 then a copy of that declaration must be filed accompanied by a copy of the decision granting § 1.47 status or, if a nonsigning person under § 1.47 has subsequently joined in a prior application, then a copy of the subsequently executed declaration must be filed. See 37 CFR 1.63(d).

NOTE: A declaration filed to complete an application must be executed, identify the specification to which it is directed, identify each inventor by full name, including the family name, and at least one given name without abbreviation together with any other given name or initial, and the residence, post office address and country of citizenship of each inventor and state whether the inventor is a sole or joint inventor. 37 CFR 1.63(a)(1)-(4).

Enclosed

Executed by

(check all applicable boxes)

inventor(s).
 legal representative of inventor(s). 37 CFR 1.42 or 1.43.
 joint inventor or person showing a proprietary interest on behalf of inventor who refused to sign or cannot be reached.
 This is the petition required by 37 CFR 1.47 and the statement required by 37 CFR 1.47 is also attached. See item 13 below for fee.

Not Enclosed.

NOTE: Where the filing is a completion in the U.S. of an International Application, or where the completion of the U.S. application contains subject matter in addition to the International Application, the application may be treated as a continuation or continuation-in-part, as the case may be, utilizing ADDED PAGE FOR NEW APPLICATION TRANSMITTAL WHERE BENEFIT OF PRIOR U.S. APPLICATION CLAIMED.

Application is made by a person authorized under 37 C.F.R. 1.41(c) on behalf of *all* the above named inventor(s).

(The declaration or oath, along with the surcharge required by 37 CFR 1.16(e), can be filed subsequently).

NOTE: It is important that all the correct inventor(s) are named for filing under 37 CFR 1.41(c) and 1.53(b).

Showing that the filing is authorized.
(not required unless called into question. 37 CFR 1.41(d))

6. Inventorship Statement

WARNING: *If the named inventors are each not the inventors of all the claims an explanation, including the ownership of the various claims at the time the last claimed invention was made, should be submitted.*

The inventorship for all the claims in this application are:

The same.
or
 Not the same. An explanation, including the ownership of the various claims at the time the last claimed invention was made,
 is submitted.
 will be submitted.

7. Language

NOTE: An application including a signed oath or declaration may be filed in a language other than English. An English translation of the non-English language application and the processing fee of \$130.00 required by 37 CFR 1.17(k) is required to be filed with the application, or within such time as may be set by the Office. 37 CFR 1.52(d).

English
 Non-English

The attached translation includes a statement that the translation is accurate. 37 C.F.R. 1.52(d).

8. Assignment

An assignment of the invention to Dairen Chemical Corporation
7th Fl., 301, SongKiang Road, Taipei, Taiwan, R.O.C.

is attached. A separate "COVER SHEET FOR ASSIGNMENT (DOCUMENT) ACCOMPANYING NEW PATENT APPLICATION" or FORM PTO 1595 is also attached.
 was filed in the parent application
 will follow.

NOTE: "If an assignment is submitted with a new application, send two separate letters-one for the application and one for the assignment" Notice of May 4, 1990 (1114 O.G. 77-78).

WARNING: A newly executed "STATEMENT UNDER 37 CFR 3.73(b)" must be filed when a continuation-in-part application is filed by an assignee. Notice of April 30, 1993, 1150 O.G. 62-64.

9. Certified Copy

Certified copy(ies) of application(s)

<u>Country</u>	<u>Appln. No.</u>	<u>Filed</u>
Taiwan	88114793	August 30, 1999

from which priority is claimed

is enclosed.
 was filed.
 will follow.

NOTE: The foreign application forming the basis for the claim for priority must be referred to in the oath or declaration. 37 CFR 1.55(a) and 1.63.

NOTE: This item is for any foreign priority for which the application being filed directly relates. If any parent U.S. application or International Application from which this application claims benefit under 35 U.S.C. 120 is itself entitled to priority from a prior foreign application, then complete item 18 on the ADDED PAGES FOR NEW APPLICATION TRANSMITTAL WHERE BENEFIT OF PRIOR U.S. APPLICATION(S) CLAIMED.

10. Fee Calculation (37 C.F.R. 1.16)

A. Regular application

CLAIMS AS FILED					
Claims	Number Filed	Basic Fee Allowance	Number Extra	Rate	Basic Fee 37 C.F.R. 1.16(a)
Total Claims (37 CFR 1.16(c))	25	- 20 =	5	x \$ 18.00	\$760.00 \$90.00
Independent Claims (37 CFR 1.16(b))	1	- 3 =	0	x \$78.00	
Multiple Dependent Claim(s), if any (37 CFR 1.16(d))			+	\$260.00	\$260.00

Amendment cancelling extra claims is enclosed.
 Amendment deleting multiple-dependencies is enclosed.
 Fee for extra claims is not being paid at this time.

NOTE: If the fees for extra claims are not paid on filing they must be paid or the claims cancelled by amendment, prior to the expiration of the time period set for response by the Patent and Trademark Office in any notice of fee deficiency. 37 CFR 1.16(d).

	Filing Fee Calculation	\$ <u>1,110.00</u>
B. <input type="checkbox"/> Design application (\$330.00—37 CFR 1.16(f))	Filing Fee Calculation	\$ _____
C. <input type="checkbox"/> Plant application (\$540.00—37 CFR 1.16(g))	Filing Fee Calculation	\$ _____

11. Small Entity Statement(s)

Statement(s) that this is a filing by a small entity under 37 CFR 1.9 and 1.27 is (are) attached.

WARNING: *"Status as a small entity must be specifically established in each application or patent in which the status is available and desired. Status as a small entity in one application or patent does not affect any other application or patent, including applications or patents which are directly or indirectly dependent upon the application or patent in which the status has been established. The refiling of an application under § 1.53 as a continuation, division, or continuation-in-part (including a continued prosecution application under § 1.53(d)), or the filing of a reissue application requires a new determination as to continued entitlement to small entity status for the continuing or reissue application. A nonprovisional application claiming benefit under 35 U.S.C. 119(e), 120, 121, or 365(c) of a prior application, or a reissue application may rely on a statement filed in the prior application or in the patent if the nonprovisional application or the reissue application includes a reference to the statement in the prior application or in the patent or includes a copy of the statement in the prior application or in the patent and status as a small entity is still proper and desired. The payment of the small entity basic statutory filing fee will be treated as such a reference for purposes of this section." 37 CFR 1.28(a)(2).*

(complete the following, if applicable)

Status as a small entity was claimed in prior application _____, filed on _____ from which benefit is being claimed for this application under:

35 U.S.C. § 119(e),
 120,
 121,
 365(c),

and which status as a small entity is still proper and desired.

A copy of the statement in the prior application is included.
Filing Fee Calculation (50% of A, B or C above) \$ 555.00

NOTE: Any excess of the full fee paid will be refunded if a small entity status is established refund request are filed within 2 months of the date of timely payment of a full fee. The two-month period is not extendable under § 1.136. 37 CFR 1.28(a).

12. Request for International-Type Search (37 C.F.R. 1.104(d))

(complete, if applicable)

Please prepare an international-type search report for this application at the time when national examination on the merits takes place.

13. Fee Payment Being Made at This Time

Not Enclosed

No filing fee is to be paid at this time.

(This and the surcharge required by 37 C.F.R. 1.16(e) can be paid subsequently.)

Enclosed

Filing fee \$ 555.00

Recording assignment
(\$40.00; 37 C.F.R. 1.21(h))
(See attached "COVER SHEET FOR
ASSIGNMENT ACCOMPANYING NEW
APPLICATION.") \$ 40.00

Petition fee for filing by other than
all the inventors or person on behalf
of the inventor where inventor
refused to sign or cannot be reached
(\$130.00; 37 C.F.R. 1.47 and 1.17(i)) \$ _____

For processing an application with a
specification in a non-English language
(\$130.00; 37 C.F.R. 1.52(d) and 1.17(k)) \$ _____

Processing and retention fee
(\$130.00; 37 C.F.R. 1.53(d) and 1.21(l)) \$ _____

Fee for international-type search report
(\$40.00; 37 C.F.R. 1.21(e)) \$ _____

NOTE: 37 CFR 1.21(l) establishes a fee for processing and retaining any application that is abandoned for failing to complete the application pursuant to 37 CFR 1.53(f) and this, as well as the changes to 37 CFR 1.53 and 1.78(a)(1), indicate that in order to obtain the benefit of a prior U.S. application, either the basic filing fee must be paid, or the processing and retention fee of § 1.21(l) must be paid, within 1 year from notification under § 53(f).

Total Fees Enclosed \$ 595.00

14. Method of Payment of Fees

Check in the amount of \$ 595.00.

Charge Account No. _____ in the amount of \$ _____.
A duplicate of this transmittal is attached.

NOTE: Fees should be itemized in such a manner that it is clear for which purpose the fees are paid. 37 CFR 1.22(b).

15. Authorization to Charge Additional Fees

WARNING: *If no fees are to be paid on filing, the following items should not be completed.*

WARNING: *Accurately count claims, especially multiple dependent claims, to avoid unexpected high charges, if extra claim charges are authorized*

The Commissioner is hereby authorized to charge the following additional fees by this paper and during the entire pendency of this application to Account No. 04-1105.

37 C.F.R. 1.16(a), (f) or (g) (filing fees)
 37 C.F.R. 1.16(b), (c) and (d) (presentation of extra claims)

NOTE: Because additional fees for excess or multiple dependent claims not paid on filing or on later presentation must only be paid or these claims cancelled by amendment prior to the expiration of the time period set for response by the PTO in any notice of fee deficiency (37 CFR 1.16(d)), it might be best not to authorize the PTO to charge additional claim fees, except possibly when dealing with amendments after final action.

37 C.F.R. 1.16(e) (surcharge for filing the basic filing fee and/or declaration on a date later than the filing date of the application)
 37 CFR 1.17(a)(1)-(5) (extension fees pursuant to § 1.136(a)).
 37 C.F.R. 1.17 (application processing fees)

NOTE: "A written request may be submitted in an application that is an authorization to treat any concurrent or future reply, requiring a petition for an extension of time under this paragraph for its timely submission, as incorporating a petition for extension of time for the appropriate length of time. An authorization to charge all required fees, fees under § 1.17, or all required extension of time fees will be treated as a constructive petition for an extension of time in any concurrent or future reply requiring a petition for an extension of time under this paragraph for its timely submission. Submission of the fee set forth in § 1.17(a) will also be treated as a constructive petition for an extension of time in any concurrent reply requiring a petition for an extension of time under this paragraph for its timely submission." 37 CFR 1.136(a)(3).

37 C.F.R. 1.18 (issue fee at or before mailing of Notice of Allowance, pursuant to 37 C.F.R. 1.311(b))

NOTE: Where an authorization to charge the issue fee to a deposit account has been filed before the mailing of a Notice of Allowance, the issue fee will be automatically charged to the deposit account at the time of mailing the notice of allowance. 37 CFR 1.311(b)).

NOTE: 37 CFR 1.28(b) requires "Notification of any change in status resulting in loss of entitlement to small entity status must be filed in the application prior to paying, or at the time of paying, . . . issue fee." From the wording of 37 CFR 1.28(b), (a) notification of change of status must be made even if the fee is paid as "other than a small entity" and (b) no notification is required if the change is to another small entity.

16. Instructions as to Overpayment

NOTE: "Amounts of twenty-five dollars or less will not be returned unless specifically requested within a reasonable time, nor will the payer be notified of such amounts; amounts over twenty-five dollars may be returned by check or, if requested, by credit to a deposit account." 37 CFR 1.26(a).

Credit Account No. 04-1105

Refund



SIGNATURE OF PRACTITIONER

Reg. No. 33,860

Peter F. Corless

(type or print name of practitioner)

Tel. No.: (617) 523-3400

Dike, Bronstein, Roberts & Cushman, LLP

130 Water Street

P.O. Address

Customer No.:

Boston, MA 02109

[X] **Incorporation by reference of added pages**

(check the following item if the application in this transmittal claims the benefit of prior U.S. application(s) (including an international application entering the U.S. stage as a continuation, divisional or C-I-P application) and complete and attach the ADDED PAGES FOR NEW APPLICATION TRANSMITTAL WHERE BENEFIT OF PRIOR U.S. APPLICATION(S) CLAIMED)

[] Plus Added Pages for New Application Transmittal Where Benefit of Prior U.S. Application(s) Claimed

Number of pages added _____

[X] Plus Added Pages for Papers Referred to in Item 4 Above

Number of pages added _____ 40

[] Plus added pages deleting names of inventor(s) named on prior application(s) who is/are no longer inventor(s) of the subject matter claimed in this application.

Number of pages added _____

[X] Plus "Assignment Cover Letter Accompanying New Application"

Number of pages added _____ 3

[] **Statement Where No Further Pages Added**

(if no further pages form a part of this Transmittal, then end this Transmittal with this page and check the following item)

[] This transmittal ends with this page.

2/22

Attorney Docket No:

BDWY Form SED-2
Small Entity
Small Business Concern

Applicant or Patentee (Inventors): (1) Shien-Chang CHEN

(發明人名字) (2) Fu-Shen LIN

Serial No. or Patent No. (if known): (3) Liang-An HSU

(申請號或專利號) (4) Pi-Fwu JANG

Filed or Issued (if known):

(申請日或公告日)

For (invention title): CATALYST FOR OXACYLATION AND USE OF THE SAME

(發明名稱)

SMALL ENTITY FORM - SMALL BUSINESS CONCERN

(VERIFIED STATEMENT/DECLARATION CLAIMING SMALL ENTITY STATUS, 37 CFR 1.9(f) and 1.27(b))

小企業聲明書 (申請人或受讓人是公司等企業時, 用此表格)

I hereby declare that I am

(若是小企業的業主, 則在下面第一個方括號內打“X”; 若是小企業的主管, 則在下面第二個方括號內打“X”)

(check one) the owner of the small business concern identified below:

an official of the small business concern empowered to act on behalf of the concern identified below:

NAME OF CONCERN: (企業名稱)

ADDRESS OF CONCERN: (地址)

I hereby declare that the above identified small business concern qualifies as a small business concern as defined in 13 CFR 121.3-18, and reproduced in 37 CFR 1.9(d), for purposes of paying reduced fees under section 41(a) and (b) of Title 35, United States Code, in that the number of employees of the concern, including those of its affiliates, does not exceed 500 persons. For purposes of this statement, (1) the number of employees of the business concern is the average over the previous fiscal year of the concern of the persons employed on a full-time, part-time or temporary basis during each of the pay periods of the fiscal year, and (2) concerns are affiliates of each other when either, directly or indirectly, one concern controls or has the power to control the other or a third party or parties controls or has the power to control both.

I hereby declare that rights under contract or law have been conveyed to and remain with the small business concern identified above with regard to the invention described in the above-identified application or patent.

If the rights held by the above identified small business concern are not exclusive, each individual, concern or organization having rights to the invention is listed below and no rights to the invention are held by any person, other than the inventor, who could not qualify as an independent inventor under 37 CFR 1.9(c) or by any concern which would not qualify as a small business concern under 37 CFR 1.9(d) or a nonprofit organization under 37 CFR 1.9(e).

FULL NAME * : (全名)

ADDRESS: (地址)

(註明一項) INDIVIDUAL SMALL BUSINESS CONCERN NONPROFIT ORGANIZATION (如果没有他人分
享本發明的權利,
(個人) (小企業) (非贏利機構) 則左邊不必填寫;

FULL NAME * : (全名)
ADDRESS: (地址) (但若有他人分享,
(註明一項) INDIVIDUAL SMALL BUSINESS CONCERN NONPROFIT ORGANIZATION 則需將他人名字、
(個人) (小企業) (非贏利機構) 地址逐項填寫, 并
需另附小企業聲明
書)

FULL NAME * : (全名)

ADDRESS: (地址)

(註明一項) INDIVIDUAL SMALL BUSINESS CONCERN NONPROFIT ORGANIZATION
(個人) (小企業) (非贏利機構)

* NOTE: Separate verified statements are required from each named person, concern or organization having rights to the invention averting to their status as small entities. (37 CFR 1.27)

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b))

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed.

Name of Person Signing: (簽字人的名字) Su-Hon LIN

Title of Person Signing: (簽字人的頭銜) President

Address of Person Signing: (簽字人的地址) 7th, Fl., 301, Songkiang Road, Taipei, Taiwan, R.O.C.

SIGNATURE



DATE

November 20, 1999

(簽名)

(日期)

TITLE: CATALYST FOR OXACYLATION AND USE OF THE SAME

FIELD OF THE INVENTION

This invention relates to a catalyst which is comprised of palladium metal as the main catalyst, tin metal or a mixture of tin and additional metal(s) as the promoter, in combination with an alkali or alkaline earth metal compound, supported on the outer surface of a porous carrier. The catalyst is used in the process for producing allyl acetate through the oxacylation of propylene, acetic acid and oxygen in a vapor phase.

BACKGROUND OF THE INVENTION

Previously, in the production of allyl acetate through the oxacylation of propylene, acetic acid and oxygen in a vapor phase, a silica carrier impregnated with palladium only was the main catalyst, and alkali or alkaline earth metal compound was used as the activator (USP 3925452). In order to get better activity from this catalyst, the oxacylation should be performed at higher temperatures. Under these conditions, the formation of carbon dioxide byproduct was increased, and the space time yield (STY, the yield per hour per liter of the catalyst) of allyl acetate was unable to be promoted even by increasing the amounts of palladium or the activator. Generally, by only using palladium as the catalyst during the oxacylation process, the space time yield of allyl acetate would not exceed 60 (g/hr/l of catalyst), and the selectivity of such a catalyst to allyl acetate would only reach 87%. In other words, most of propylene reactant was burned into carbon dioxide or converted into other byproducts. Apparently, when only palladium and the activator were used as the catalyst, the catalytic ability of the catalyst was rather low, and waste resulted from the complete burning of propylene into carbon dioxide, which adversely influenced the industrial process.

In order to improve on this drawback, in the preparation process of the

catalyst for oxacylation reaction, other metals were added so as to increase the activity and selectivity of the catalyst (USP 3917676). Therefore, most of the compositions of the catalysts were comprised of, not only palladium, the main catalyst, and alkali or alkaline earth metal, the activator, but also other metals, as the promoters. For example, along with catalysts with the combination of the main catalyst palladium and the promoters of potassium, bismuth and barium (USP 4571431), the combination of the main catalyst palladium and the promoters of copper, lead, ruthenium and rhenium (EP 0361484), etc. have been disclosed. Among these, a catalyst with the combination of the main catalyst palladium and the promoter copper exhibited higher activity and selectivity (USP 5011980).

For the purpose of high catalytic activity and high catalyst selectivity, besides the addition of other metals as the promoter in preparing the catalyst, a certain amount of water as a diluent should be added into the feeding materials of propylene, acetic acid and oxygen for performing the oxacylation reaction, in the traditional process of producing allyl acetate. If moisture content was under a certain ratio, the catalytic activity and life of the catalyst could not be retained and would deteriorate rapidly. Usually, the addition of the water diluent would limit the yield of the final product. Further, the final product, allyl acetate, should be purified after the oxacylation, which would result in consuming and wasting energy of the whole process, and the economical effect could not be attained.

In the presence of the catalyst produced in this invention, while no water is added into the reactant materials for the oxacylation process, or only a small amount of water is added in accordance with the requirement of the process, not only will the catalytic activity and life of the catalyst be retained and not deteriorate, but a high catalytic activity and high selectivity will be attained. Therefore, the energy consumed and wasted resulting from the addition of

water can be avoided, and the economical effect of the oxacylation process can be greatly increased.

SUMMARY OF THE INVENTION

This invention relates to a catalyst which is comprised of palladium metal as the main catalyst, tin metal or a mixture of tin and additional metal(s) as the promoter, in combination with an alkali or alkaline earth metal compound, supported on the outer surface of a porous carrier. The catalyst is used in the process for producing allyl acetate through the oxacylation of propylene, acetic acid and oxygen in a vapor phase. The catalyst of the present invention exhibits high catalytic activity, high catalytic selectivity and high catalytic life, which greatly increases the economic utility of the oxacylation process.

DETAILED DESCRIPTION OF THE INVENTION

The porous carriers which are suitable for preparing the catalyst for oxacylation of the present invention are alumina, silica gel, silica, active carbon, silicon carbide, diatomaceous earth, pumice and the like, while among these, silica and alumina are preferable.

The main catalyst metal of the catalyst for oxacylation of the present invention is palladium; the metal content thereof, based on the weight of the carrier, is 0.1 to 5.0 weight %, preferably 0.3 to 1.5 weight %. The promoter metal of the catalyst for oxacylation of the present invention is tin or a mixture of tin and additional metal(s) selected from the group consisting of gold, copper, cadmium, bismuth, cerium and a mixture thereof, while among these, a mixture of tin and gold is preferable; the metal content thereof, based on the weight of the carrier, is 0.01 to 5.0 weight %, preferably 0.02 to 1.0 weight %. The activator of the catalyst for oxacylation of the present invention is an alkali or alkaline earth metal compound, the examples thereof being the hydroxides, acetates, nitrates and bicarbonates of potassium, sodium, cesium, magnesium,

barium and the like, while among these, potassium salts are preferable, and potassium acetate is even more preferable. The content thereof, based on the weight of the carrier, is 1 to 15 weight %, preferably 4 to 10 weight %.

Traditionally, the preparation method of the catalyst for oxacylation was essentially comprised of the following steps: (1) a carrier was impregnated with an aqueous solution of soluble palladium ions and metal ions of the promoter; (2) the impregnated carrier was immersed in an alkali solution, so that the soluble palladium ions and metal ions of the promoter were precipitated on the surface layer of the carrier and formed into insoluble oxidative state palladium and promoter metal; (3) the treated carrier was washed with water to remove soluble ions produced during the precipitation; (4) the oxidative state palladium and promoter metal supported on the treated carrier were then reduced to the metallic state; (5) the reduced carrier in (4) was impregnated with a solution of an alkali or alkaline earth metal compound; and (6) the impregnated carrier in (5) was dried. The term "oxidative state" used herein according to the present invention means a metal in a cationic state, for example, oxidative state palladium means Pd^{2+} .

The catalyst for oxacylation of the present invention is prepared mainly in accordance with the traditional method. After the oxidative state palladium and promoter metal are supported on the surface of the porous carrier, this not-reduced yet catalyst is placed in a reactor and the reducing step is performed under suitable reductive conditions using gaseous or liquid reducing agents. The examples of the reducing agents are amines, carbon monoxide, hydrogen, alkene, aldehydes and hydrazines. When gaseous reducing agents are used, it is preferable to dilute the gaseous reducing agent with inert gas (such as nitrogen gas) The amount of the reducing agent used depends on the amounts of the palladium and the promoter metal, the equivalents used thereof usually being at least 1 to 1.5 times of the equivalents required to reduce the

catalyst. If necessary, more reducing agent can be used. After the reducing process, the reduced catalyst is washed with deionized water until the chloride ions are completely removed and then dried. After drying, the reduced catalyst is impregnated with an aqueous solution containing an alkali or alkaline earth metal compound. Finally, the catalyst is dried at a temperature between 80 to 150°C.

A certain amount of the above prepared catalyst for oxacylation is placed in a reacting tube with an inner diameter of 20mm and a length of 2.0 m. Under a specific pressure at the inlet of the reacting tube, the reactant feeding gases are introduced into the tube at a reacting temperature set according to the activity of the catalyst. These reactant feedings comprise propylene, nitrogen, acetic acid, oxygen and water, wherein the content of propylene is 20 to 50 volume %; the content of nitrogen is 20 to 60 volume %; the content of acetic acid is 5 to 25 volume %; the content of oxygen is 5 to 10 volume %; and the content of water is 0 to 15 volume %, preferably 0 to 10 volume %. The catalyst for oxacylation of the present invention is characterized in that, the catalytic activity and life of the catalyst can be retained and will not deteriorate while no water is added into the reactant composition, or if only a small amount of water is added in the oxacylation process.

The operation temperature of the above oxacylation process is in the range of 100°C to 250°C, preferably 140°C to 200°C; the operation pressure is in the range of 0 to 15 kg/cm²·g, preferable 5 to 10 kg/cm²·g.

The yield of allyl acetate is determined by analyzing the composition at the exit when the oxacylation process is carried out for a definite time.

Generally, the selection of a catalyst in the industry is based on the catalytic activity (STY). The catalytic activity can be calculated basically

according to the following formula:

The activity of a catalyst:

$$\text{STY(space time yield)} = \frac{\text{weight of allyl acetates produced(g)}}{\text{volume(l) of catalyst} \times \text{sampling time(hr)}}$$

The selectivity of a catalyst:

$$\text{Allyl acetate selectivity} = \frac{\text{moles of allyl acetate produced}}{\text{moles of allyl acetate produced} + \frac{1}{3} \text{ moles of CO}_2 \text{ produced}}$$

It is confirmed from the evaluation of the catalytic activity in the oxacylation process' practical application that the catalyst for oxacylation prepared in accordance with the present invention not only provides higher activity of the whole oxacylation reaction of propylene, acetic acid and oxygen, but also prolongs its own life. That is, compared to the conventional catalysts, the catalyst of the present invention is able to yield more allyl acetate per unit volume of catalyst in the reactor and per unit time, while no water or only a small amount of water is added in the oxacylation process, and the conditions of the oxacylation reaction (such as pressure, temperature, oxygen concentration) remain constant. Thus, the energy wasted resulting from the traditional process by using large amounts of water can be reduced. Moreover, if the productive yield remains constant, not only can the reacting temperature be decreased, but also the selectivity of the reaction can be higher, which leads to less production of carbon dioxide and less product loss during the removal of carbon dioxide. Thus, the unit raw material consumption will be lower. This is beneficial to the industrial production of allyl acetate.

The present invention will be further described with reference to the following Examples and Comparative Examples, but the scope of the present invention is by no means limited.

Example 1

The carrier employed in this Example was a porous carrier of alumina/silica with an outer diameter of 5 mm and available from SUD-CHEMIE AG. This carrier had a surface area of 100 to 120 m²/g, a pore volume of 0.7 to 0.9 ml/g and a bulk density of 600 g/l. The metal component-supporting catalyst was prepared according to the following steps:

Step 1): An aqueous Na₂PdCl₄ solution with weight of 2.2 kg containing 15 weight % of palladium was added into a mixture of an aqueous SnCl₂ solution with weight of 0.5 kg containing 15 weight % of tin and an aqueous HAuCl₄ solution with weight of 0.5 kg containing 30 weight % of gold. The mixture was then diluted with deionized water till total volume was 37.2 liters. One-hundred liters of alumina/silica carrier was placed in an impregnating tank with rotation rate of 24 turns per minute. The mixture was added into the tank rapidly.

Step 2): Hot air was passed through to dry the carrier. The temperature of the hot air was lower than 120°C

Step 3): Twenty-eight weight % of NaOH solution (about 60 kg) was added to the dried catalyst. The originally soluble chloride state palladium, tin and gold were transformed into insoluble hydroxide state palladium, tin and gold.

Step 4): The impregnated catalyst carrier after drying was placed in a reducing reactor. The reducing gases were passed into the reactor, wherein the reducing gases could be diluted with other inert gases. The hydroxide state metal catalyst was reduced into a metallic state catalyst.

Step 5): The above catalyst was washed to remove chloride ions until the catalyst was free of chloride ions.

Step 6): The catalyst carrier was dried as in step 2).

Step 7): An adequate amount of potassium acetate was added into the dried catalyst carrier, so that each liter of the catalyst contained 30 g weight of potassium acetate.

Step 8): The catalyst carrier was dried as in step 2).

After the above steps, a catalyst containing 3.3 g/l of palladium, 0.75 g/l of tin, 1.5 g/l of gold and 30 g/l of potassium acetate was obtained, wherein all palladium, tin and gold were well distributed on the surface of the carrier.

Four hundred and fifty milliliters of the catalyst thus obtained was charged into a reacting tube with an inner diameter of 20 mm and a length of 2.0 m. Under a pressure of 7 kg/cm² (gauge pressure) at the inlet of the reactor, the reacting gaseous mixture was introduced into the reactor at a temperature of 140°C. The gaseous mixture was comprised of 41 volume % of propylene, 43 volume % of nitrogen gas, 10 volume % of acetic acid and 6 volume % of oxygen. When the composition at the exit was analyzed in a definite time, the activity and the selectivity of the catalyst were calculated. The results are listed in Table 1.

When the activity and the selectivity of the catalyst were evaluated, the crude product at the exit of the reactor was cooled with chilled water, and the composition was analyzed by Shimadzu Gas Chromatography. The flow rate of the gases was determined by Shinagawa Dry Gas Meter.

Example 2

The catalyst was prepared by the same method as in Example 1 except that, in step 1), an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium, an aqueous SnCl_2 solution with weight of 0.5 kg containing 15 weight % of tin and an aqueous CuCl_2 solution with weight of 0.5 kg containing 15 weight % of copper were prepared.

This catalyst was evaluated by the same method as in Example 1, and the results are listed in Table 1.

Example 3

The catalyst was prepared by the same method as in Example 1 except that, in step 1), an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium and an aqueous SnCl_2 solution with weight of 0.5 kg containing 15 weight % of tin were prepared.

This catalyst was evaluated by the same method as in Example 1, and the results are listed in Table 1.

Example 4

The catalyst was prepared exactly by the same method as in Example 1, i.e., an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium, an aqueous SnCl_2 solution with weight of 0.5 kg containing 15 weight % of tin and an aqueous HAuCl_4 solution with weight of 0.5 kg containing 30 weight % of gold were prepared.

This catalyst was evaluated by the same method as in Example 1 except that, the gaseous mixture for performing oxacylation reaction was comprised of 41 volume % of propylene, 37 volume % of nitrogen gas, 9 volume % of acetic acid, 6 volume % of oxygen and 7 volume % of water, and the results are listed in Table 1.

Comparative Example 1

The catalyst was prepared by the same method as in Example 1 except that, in step 1), an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium and an aqueous HAuCl_4 solution with weight of 0.5 kg containing 30 weight % of gold were prepared.

This catalyst was evaluated by the same method as in Example 4, and the results are listed in Table 1.

Comparative Example 2

The catalyst was prepared by the same method as in Example 1 except that, in step 1), an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium and an aqueous CuCl_2 solution with weight of 0.5 kg containing 14.6 weight % of copper were prepared.

This catalyst was evaluated by the same method as in Example 4, and the results are listed in Table 1.

Comparative Example 3

The catalyst was prepared by the same method as in Example 1 except that, in step 1), an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium and an aqueous HAuCl_4 solution with weight of 0.5 kg containing 30 weight % of gold were prepared.

This catalyst was evaluated by the same method as in Example 1, and the results are listed in Table 1.

Comparative Example 4

The catalyst was prepared by the same method as in Example 1 except that, in step 1), an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium and an aqueous CuCl_2 solution with weight of 0.5 kg containing 14.6 weight % of copper were prepared.

This catalyst was evaluated by the same method as in Example 1, and the

results are listed in Table 1.

Comparative Example 5

The catalyst was prepared by the same method as in Example 1 except that, in step 1), an aqueous Na_2PdCl_4 solution with weight of 2.2 kg containing 15 weight % of palladium, an aqueous HAuCl_4 solution with weight of 0.5 kg containing 30 weight % of gold and an aqueous CuCl_2 solution with weight of 0.67 kg containing 15 weight % of copper were prepared.

This catalyst was evaluated by the same method as in Example 1, and the results are listed in Table 1.

Table 1

Item	STY-1(g/l/hr)	STY-2(g/l/hr)	Relative Ratio	Selectivity(%)
Example 1	400	355	0.887	94.8
Example 2	323	204	0.632	91.6
Example 3	350	225	0.643	92.5
Example 4	410	368	0.898	95.1
Comparative Example 1	400	312	0.780	91.9
Comparative Example 2	420	353	0.840	94.5
Comparative Example 3	375	122	0.325	92.8
Comparative Example 4	450	162	0.360	95.1
Comparative Example 5	385	200	0.519	94.7

Note: 1. STY-1: Space time yield of allyl acetate after the oxacylation reaction was carried out for 6 hours.

2. STY-2: Space time yield of allyl acetate after the oxacylation reaction was carried out for 120 hours.

3. Relative ratio: The catalytic activity deteriorating ratio of STY-2 to STY-1.

It is seen clearly from the above Examples and Comparative Examples that, the activity of the traditional catalyst (see Comparative Examples 1 to 5, wherein gold or copper was used as the catalysis promoter) deteriorates rapidly when water is not added during the oxacylation process. As to the catalyst for oxacylation produced in the present invention, whether only tin, or the mixture of tin/gold or tin/copper is used as the promoter, superior catalytic activity deteriorating ratios are obtained when water is not added during the oxacylation process.

Therefore, in the presence of the catalyst produced in this invention, while no water is added into the reactant materials for the oxacylation process, or if only a small amount of water is added in accordance with the requirement of the process, not only can the catalytic activity and life of the catalyst be retained and not deteriorate, but high catalytic activity and high selectivity will be attained. Therefore, the energy consumed and wasted resulting from the addition of water can be avoided, and the economical effect of the oxacylation process can be greatly increased.

What is claimed is:

1. A catalyst for oxacylation, which comprises palladium metal as the main catalyst, tin metal or a mixture of tin and additional metal(s) as the promoter, in combination with an alkali or alkaline earth metal compound, supported on the outer surface of a porous carrier, and being used in the process for producing allyl acetate.
2. The catalyst according to claim 1, which further comprises metal(s) selected from the group consisting of gold, copper, cadmium, bismuth, cerium and a mixture thereof as the additional promoter.
3. The catalyst according to claim 1, wherein the content of said main catalyst,

palladium metal, based on the weight of said porous carrier, is in the range of 0.1 to 5.0 weight %.

4. The catalyst according to claim 3, wherein the content of said main catalyst, palladium metal, based on the weight of said porous carrier, is in the range of 0.3 to 1.5 weight %.
5. The catalyst according to claim 1, wherein the content of said promoter, tin metal, based on the weight of said porous carrier, is in the range of 0.01 to 5.0 weight %.
6. The catalyst according to claim 5, wherein the content of said promoter, tin metal, based on the weight of said porous carrier, is in the range of 0.02 to 1.0 weight %.
7. The catalyst according to claim 2, wherein the total content of said promoter, tin metal and additional metal(s), based on the weight of said porous carrier, is in the range of 0.01 to 5.0% by weight.
8. The catalyst according to claim 7, wherein the total content of said promoter, tin metal additional promoter metal(s), based on the weight of said porous carrier, is in the range of 0.02 to 1 0% by weight.
9. The catalyst according to claim 1, wherein the content of said alkali or alkaline earth metal compound, based on the weight of said porous carrier, is in the range of 1 to 15 weight %.
10. The catalyst according to claim 9, wherein the content of said alkali or alkaline earth metal compound, based on the weight of said porous carrier, is in the range of 4 to 10 weight %.
11. The catalyst according to claims 7 or 8, wherein said additional promoter metal is gold.
12. The catalyst according to claims 7 or 8, wherein said additional promoter metal is copper.
13. The catalyst according to claims 7 or 8, wherein said additional promoter metal is selected from the group consisting of cadmium, bismuth and cerium.

14. The catalyst according to claim 1, wherein said alkali or alkaline earth metal compounds are the hydroxides, acetates, nitrates and bicarbonates of potassium, sodium, cesium, magnesium and barium.
15. The catalyst according to claim 14, wherein said alkali or alkaline earth metal compounds are the hydroxide, acetate, nitrate and bicarbonate of potassium.
16. The catalyst according to claim 1, wherein said porous carrier is selected from the group consisting of alumina, silica gel, silica, active carbon, silicon carbide, diatomaceous earth, pumice and a mixture thereof.
17. The catalyst according to claim 1, wherein said process for producing allyl acetate is carried out through the oxacylation of propylene, acetic acid, oxygen and water in a vapor phase.
18. The catalyst according to claim 17, wherein the content of water is in the range of 0 to 15 volume %, based on total amount of the reacting gases.
19. The catalyst according to claim 18, wherein the content of water is in the range of 0 to 10 volume %, based on total amount of the reacting gases.
20. A method for preparing the catalyst according to claim 1, which comprises: (a) impregnating a porous carrier with a solution containing palladium and promoter metal(s) in oxidative states, then reducing the metals from an oxidative state into metallic state; (b) impregnating said metallic state metals-supporting carrier with a solution of alkali or alkaline earth metal compounds, then drying it.
21. The method according to claim 20, wherein the reduction reaction for reducing the metals from an oxidative state into a metallic state is carried out in a liquid phase, and the reducing agent used is selected from the group consisting of amines, aldehydes and hydrazines.
22. The method according to claim 20, wherein said reduction reaction for reducing the metals from an oxidative state into a metallic state is carried out in a vapor phase, and the reducing agent used is selected from the group consisting of carbon monoxide, hydrogen and alkene.

ABSTRACT

This invention relates to a catalyst which comprises palladium metal as the main catalyst, tin metal or a mixture of tin and additional metals as the promoter, in combination with an alkali or alkaline earth metal compound, supported on the outer surface of a porous carrier. The catalyst is used in the process for producing allyl acetate through the oxacylation of propylene, acetic acid and oxygen in a vapor phase. The catalyst of the present invention exhibits high catalytic activity, high catalytic selectivity and high catalytic life, which greatly increases the economic utility of the oxacylation process.

21222

Docket No. _____

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

CATALYST FOR OXACYLATION AND USE OF THE SAME

the specification of which

(check one)

is attached hereto.

was filed on _____ as United States Application No. or PCT International

Application Number _____

and was amended on _____

(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed _____

88114793 (Number)	Taiwan R. O.C. (Country)	August 30, 1999 (Day/Month/Year Filed)	<input type="checkbox"/>
_____	_____	_____	<input type="checkbox"/>
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_____	_____	_____	<input type="checkbox"/>

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112, I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C. F. R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)

(Filing Date)

(Status)

(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)

(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)

(patented, pending, abandoned)

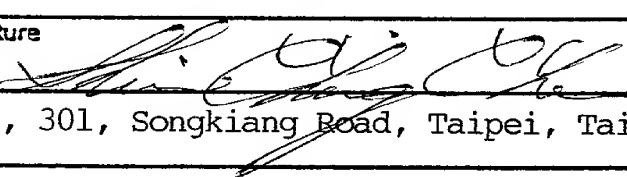
I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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